Setting the Standard for Atomic-Scale Measurements

Atomic nuclei in a crystal can absorb and emit photons with an extremely well-defined wavelength. This wavelength is measured with a relative uncertainty of 0.19 ppm by using Bragg scattering at near 90° from a reference Si crystal.

The definition of the meter as a length standard evolved with our growing knowledge of the basic laws of physics. In 1799, a platinum bar made as close as possible to the one ten-millionth part of the polar quadrant of the earth passing through Paris was defined as one meter and was deposited as a reference in the French National Archive. Replaced in 1899 by an "International Prototype Meter" in the form of an alloy bar of platinum (90%) and iridium (10%), it served as the length standard until 1960, when the wavelength of the orange line from a ^{86}Kr standard lamp took over this role. Since 1983, the meter has been defined as the distance traveled by light in 1/299792458 of a second. Here, the speed of light c is converted to a constant without uncertainty, and the wavelength standard λ_S is established via measurement of the frequency f_S of an iodine stabilized He-Ne laser such that $\lambda_S = c/f_S$. This definition provides a highly accurate length standard due to the narrowness and stability (a few parts in 10^{11}) of the laser radiation line and the ability to measure laser frequencies with respect to the ^{133}Cs standard.

However, the wavelength of the He-Ne laser is large in comparison to interatomic distances, which are typically ten thousand times smaller. Therefore, a secondary length standard was created that is better suited for distances on the atomic scale.

The combination of a Michelson visible-light interferometer and a Bonse-Hart x-ray interferometer permits determination of the lattice constant of high-quality silicon single crystals in terms of the wavelength of the He-Ne laser with a relative uncertainty of 0.029 ppm. The lattice constant of silicon $a = 5.43102088(16) \times 10^{-10}$ m at 22.500ºC is the most accurate length standard in use for interatomic measurements.

However, to reproduce its value even with a moderate relative uncertainty of 1 ppm is not effortless. Chemical purity, crystal perfection, absolute temperature, and pressure have to be precisely controlled. This is possible in well-equipped laboratories but is not a trivial task. Besides, to gauge interatomic distances, one eventually needs radiation with a well-defined wavelength rather than a well-defined length standard. An easily reproducible wavelength standard for the atomic scale is clearly desirable.

Certain nuclear excited states are known for their extremely narrow line width. If such an isotope is embedded in a solid, Mössbauer radiation with a similarly narrow spectral line width is emitted. The Mössbauer effect, which was discovered in 1958, prevents line broadening from lattice vibrations and renders the precise and stable wavelength of Mössbauer radiation. An excellent example is radiation emitted in the transition of ^{57}Fe nuclei from the first excited state to the ground state. The energy of the Mössbauer radiation is $E_M \cong 14.4$ keV, and the corresponding wavelength of $\lambda_M \cong 0.86 \times 10^{-10}$ m is perfectly suited to be used as a wavelength standard for atomic scales.

The spectral line width Γ of the Mössbauer radiation is related to the lifetime of the excited state $\tau = 141$ ns by the relation $\Gamma\tau = \hbar$ and is only

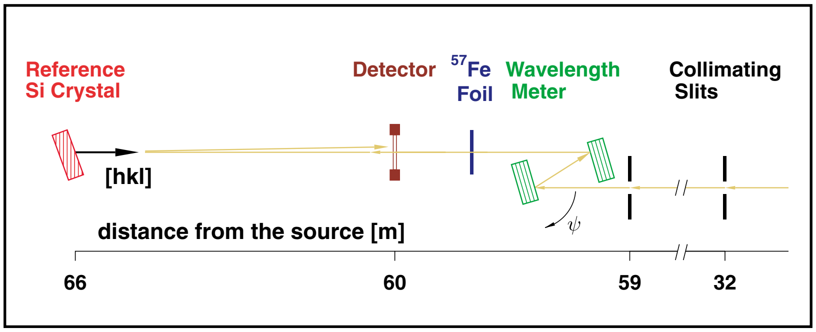

FIG. 1. A schematic of the experiment.

absolute value of the wavelength. There are at least two options. One could measure a Mössbauer wavelength directly in terms of the wavelength of the He-Ne laser. Another, less direct, way is to measure the Mössbauer wavelength with respect to the silicon lattice constant—i.e., silicon is used as a transfer standard. We have chosen the second approach.

4.7×10^{-9} eV. Thus, the relative energy and wavelength uncertainty of the Mössbauer radiation is $\Gamma/E_M \cong 3 \times 10^{-13}$. A potential source of line broadening originates in interaction of the nuclei with the environment, e.g., the electrons of the atomic core. These so-called hyperfine interactions may deteriorate the spectral purity of the Mössbauer radiation by a factor of 100. Thus, even if nothing is known about the hyperfine interactions, the relative uncertainty and reproducibility is still $\cong 10^{-11}$. The outstandingly small uncertainty is a gift of nature and makes Mössbauer radiation an attractive wavelength standard.

In practical use of a wavelength standard, reproducibility is also essential. Ideally, one needs strong and well-collimated sources of Mössbauer radiation that are easily available. Here, the recent advent of third-generation synchrotron radiation sources comes to the rescue. Any iron-containing substance exposed to synchrotron radiation in the 14.4 keV spectral range becomes a strong source of Mössbauer radiation, while the high brightness of the incident beam is preserved. The availability of such sources has stimulated our attempts to establish Mössbauer radiation as the length standard in the x-ray regime.

After realizing the potential of the Mössbauer wavelength standard, an important first step leading to practical use is the precise measurement of the

The angular divergence of the synchrotron radiation was reduced to 4 μrad with two micro-meter slits placed 27 m apart (Fig. 1). A particular wavelength of the x-rays was selected by the angle position ψ of a wavelength meter, which is essentially a very stable monochromator with meV energy resolution. The rotation angle ψ was calibrated in the units of the well-known lattice constant of a reference silicon crystal, which served as the transfer standard. Temperature, pressure, and the crystal composition were well controlled. The calibration was performed by observing nearly exact Bragg backscattering from different sets of atomiplanes

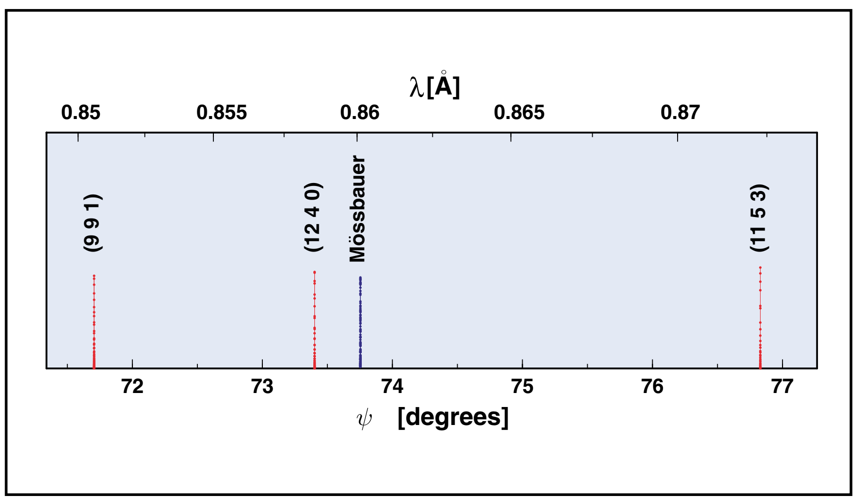

FIG. 2. Back-reflected and Mössbauer radiation in the detector vs the rotation angle ψ of the wavelength meter.

of the reference crystal. After the wavelength meter was calibrated, the Mössbauer wavelength could be measured. An iron foil was installed in the beam. At some angle of the wavelength meter, the wavelength of the transmitted radiation coincides with λ_M (Fig. 2). At precisely this angle, resonant excitation of the

^{57}Fe nuclei occurs, and photons are re-emitted with a delay of typically 140 nanoseconds relative to the incident synchrotron radiation pulse. The delayed emission permits discrimination and provides a clear signal of nuclear resonant excitation. As a result, the wavelength λ_M was measured to be $0.86025474 \times 10^{-10}$ m with an uncertainty of 0.19 ppm.

The first step to establish Mössbauer radiation as a wavelength standard in the x-ray regime has been successfully completed. At this early stage, independent confirmation of our results is necessary, but we are confident that the precision can be further improved in similar experiments. When the accuracy of the transfer standard is reached, one probably has to convert to more direct methods. Most exciting would be the direct measurement of the Mössbauer wavelength in terms of the wavelength of the He-Ne laser. We are at the beginning of a long road. It is difficult to predict where it will end, but we are certain that many benefits await us on the roadside to make the effort worthwhile.

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